Study on Radioactivity Components, Water Quality and Microstructure Characteristic of Volcano Ash as Geopolymer Artificial Aggregate

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The assessments of radioactivity, water quality testing and microstructure characteristic of volcano ash have been examined. The measurement of the ^{226}Ra, ^{232}Th, ^{40}K, and ^{238}U were carried out using radioactive concentration value. The results showed that ^{226}Ra = 39 Bq/kg, ^{232}Th = 36 Bq/kg, ^{40}K = 337 Bq/kg which are within acceptable limit for construction building. The radium equivalent activity for volcano ash is 116 ± 1 Bq/kg, means the exposure of γ radiation is safe. Water from volcano ash may affected the aquatic ecosystem and human health which is not safe to be used as effluent due to high zinc, cadmium, lead, cyanide and sulphide and exceed the allowable limits. The mean particle size of the volcano ash was 121 μm. This volcano ash is almost dominated by quartz phase. SEM analysis showed that volcano ash had a plate-like structure similar to kaolin. The FTIR adsorption band showed the OH-, H-O-H, Si-O, Al-OH, and Si-O-Si and Si-O-Al vibrations appeared in this volcano ash. The average total percentage of weight loss after being heated to 1000°C was 15.85%.

Keywords: radioactivity, water quality, geopolymer, XRF, XRD

A six-year-old volcano mud from Sidoarjo, East Java, Indonesia started to form on May 29, 2006 with an eruption site known as LUSI (LU-lumpur, SI-Sidoarjo) and is still active until today [1]. Large quantities of volcano ash which started to erupt caused by drilling of the gas exploration well in the Porong area, Sidoarjo, East Java [2-4] or due to the Yogyakarta earthquake that occurred at 05:54 am on the 27th May 2006 [3, 5]. This eruption has begun when it surfaced from the bowels of the earth and impacted an area of almost 3 square miles to a depth of 65 feet and thirty thousand people have been displaced which cost Indonesia $3.7 billion in damages and damage control [1, 5, 6].

Numerous efforts to stop the eruption of volcano mud have failed. However, the mud flow is now manageable. After peaking at 180.000 cubic meters per day in early 2007, the rate has tapered to 10.000 cubic meters per day [7, 8]. A system of 6.0m to 7.0m high earthen dikes encloses some 700 hectares of ponds where mud and water is collected and then pumped into the Porong River as shown in figure 1, where it is adding to a natural delta downstream [2, 6, 7]. The impact on the Porong has been minimal, given that it historically carried heavy sediment loads from magmatic volcanoes upstream. In this case, the effect on Porong needs to be studied by analyzing the water quality tests. Analysis also needs to be taken on examining its impact on human health and the environment by radioactivity tests.

In recent years, the recycling of industrial waste or by-products containing Technologically Enhanced Natural Occurring Radioactive Materials (TENORM) is extensively used in the construction industry. The use of fly ash, coal slag, red mud and other industrial byproducts in building materials and the increment in radiation exposure from these materials it has been of concerned for several years. Geopolymer developed by Davidovits [9-11] introduce new technology in a novel family of building materials. Geopolymer does not utilize any Portland cement in its production. Geopolymer is formed by alkali activation of aluminosilicate raw materials from industrial waste and by-product material like fly ash, metakaolin, blast furnace slag, rice husk ash and palm ash. The source materials for geopolymer should be rich in silicon (Si) and aluminium (Al). The natural minerals such as kaolin and clays also can be used as source materials [9, 12, 13]. However, the choice of the source materials for making geopolymers should be take into account the availability, cost, type of application and demand in the market [13]. So, the idea to

Fig. 1. Water from volcano mud is pumped into the Porong river

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utilize the volcano ash from Sidoarjo eruption site as raw material in geopolymer has been created as an alternative to convert this material into the building materials due to their high contents of Si and Al.

Hazama and Shizuma [14] have studied the impact of radioactivity to the environment of volcano ash from Sidoarjo, Indonesia representing the volcano ash on year 2009 and found that the activity concentrations were within the acceptable limits to be used in the construction building to any significant radiation exposure to the occupants. However, the continuous testing on the radiation and water quality need to be done since the eruption of volcano ash is unstoppable until now. Also, the chemical soil charge coming from geochemical processes before the eruption, may contaminate groundwater with toxic transition metal cations and anions [15-21].

This paper asses the contents of ^{226}Ra, ^{232}Th, ^{40}K, and ^{238}U whether is still in the acceptable limit after 6 years disaster. The current water quality testing also has determined and compared to the Environmental Quality (Sewage & Industrial Effluent) Regulations 1979 [22].

The microstructure characteristic of volcano ash also have been presented in this paper including Particle Size Analyzer (PSA), *X-ray fluorescence* (*XRF*), x-ray diffraction (XRD), scanning electron microscopy (SEM), Fourier Transform Infrared Spectroscopy (FTIR), Thermogravimetric analysis (TGA) and Differential Thermogravimetric (DTG).

Experimental part

Sampling

The present study was conducted on the volcano ash collected from the eruption sites called the LUSI near Sidoarjo, East Java, Indonesia. The material was then transferred into sealed container. The mud was medium gray in colour as shown in figure 2. The water collected at eruption site was also sealed in the bottle for water quality testing. The volcano ash collected was then dried in the oven at 105°C for 24 h. Then, the dried volcano ash was crushed and blended to small particle size to form as volcano ash.

Fig. 2. The fresh volcano ash collected from Sidoarjo, Indonesia in 2012

Experimental Testing

The prepared samples of volcano ash was sent to the Malaysian Nuclear Agency (Nuclear Malaysia) to determine the radioactive component in the volcano ash samples either still in the required limitation. The radioactive analysis was done for the Uranium (U-238), Thorium (Th-232), Radium (Ra-226, Ra-228) and Kalium (K-40) to find their level of radioactivity using Neutron Activation Analysis (NAA) and in-house method, RAS-TM-01.

The water collected from eruption site of volcano ash was tested for water quality testing to check their parameter content and limitation. One litre sample of water volcano ash was analysed to check the contents cations of Mercury (Hg^{2+}), Tin (Sn^{2+}), Cadmium (Cd^{2+}), Arsenic (As^{3+}), Lead (Pb^{2+}), Chromium (Cr^{3+}), Copper (Cu^{2+}), Manganese (Mn^{2+}), Nickle (Ni^{2+}), Zinc (Zn^{2+}), Iron ($Fe^{2/3+}$), Boron (B^{3+}),and anions Cyanide (CN^{-}) and Sulphide (S^{2-}). The method used was based on Standard Methods for the Examination of Water and Wastewater (APHA) and HACH DR 2800 Spectrophotometer Procedures Manual.

The microstructure characterization of volcano ash samples were analyzed to find the characteristic of raw material. The particle size analysis (PSA) has been done to find the mean size of volcano ash particle. *X-ray fluorescence* (*XRF*) was used to measure the chemical composition of volcano ash and the details are shown in table 1. X-ray diffraction (XRD) patterns were performed and this test was held to investigate the phase analysis of the original volcano ash. Scanning electron microscopy (SEM) was performed to investigate the microstructure and shape of the original volcano ash. Fourier transform infrared spectroscopy (FTIR) was performed to determine the bonding between the particles in volcano ash. All samples were prepared in powder form for testing purpose. Thermogravimetric analysis (TGA) and Differential thermogravimetric (DTG) were performed to find the weight loss as a function of temperature under a controlled atmosphere.

Table 1
CHEMICAL COMPOSITION OF VOLCANO ASH

Component	Volcano ash (%)
Al_2O_3	14.60
SiO_2	40.00
K_2O	4.28
TiO_2	1.75
Fe_2O_3	23.25
CaO	5.46
MnO	0.34
SO_3	0.88
V_2O_5	0.064

Results and discussions

Environmental Assessment of Volcano ash

Radioactivity Level

Based on European Commission Guidance, the document proposes the introduction of an activity concentration index (*I*) to be used as the guideline of the safety requirement of building materials. The equation on finding activity concentration index (I) is shown in equation (1)[14]:

$$I = \frac{A_{Th}}{300\ (\frac{Bq}{kg})} + \frac{A_{Ra}}{200\ (\frac{Bq}{kg})} + \frac{A_K}{3000\ (\frac{Bq}{kg})} \quad (1)$$

Where A_{Th}, A_{Ra}, and A_K are the thorium, radium and potassium activity concentrations (Bq/kg). These results were compared with the study carried out by Hazama and Shizuma [14] to present the characterization of volcano ash on year 2009 and now 2012 (using the same sources of volcano ash from Sidoarjo, Indonesia) and summarized in table 2. However, the radionuclide concentration of our results showed lower than the world average for building materials (^{226}Ra = 50 Bq/kg, ^{232}Th = 50 Bq/kg, ^{40}K = 500 Bq/kg). Similarly, this volcano ash also showed lower population weighted averages in soils compared to the standard from The United Nations Disaster Assessment and Coordination (UNDAC, 2006) [23]; ^{238}U = 33Bq/kg, ^{232}Th = 45 Bq/kg, ^{40}K = 420 Bq/kg) [24].

Table 3 showed the relationship between the activity concentration index (*I*) and received dose per year as stated by European Commission Radiation Protection

Volcano ash Sample	Tested sample, 2012	Tested sample by Hazama and Shizuma, 2009 [14]	
		A	B
^{226}Ra (Bq/kg)	39±7.15	49±8	61±10
^{232}Th (Bq/kg)	36	48±8	86±10
^{40}K (Bq/kg)	337.38±58.26	740±140	580±130
^{238}U (Bq/kg)	20	-	-
Activity Index (*I*)	0.43±0.03	0.65±0.07	0.83±0.08

Table 2
RESULTS OF RADIONUCLIDE CONCENTRATION AND ACTIVITY INDEX (*I*) COMPARED WITH HAZAMA AND SHIZUMA [14] STUDY

Dose criterion	< 0.3 mSv	< 1 mSv
Bulk Materials, eg. Concrete	$I \leq 0.5$	$I \leq 1$
Superficial and other materials with restricted use : tiles, boards, etc.	$I \leq 2$	$I \leq 6$

Table 3
RELATIONSHIP BETWEEN THE ACTIVITY CONCENTRATION INDEX (*I*) AND RECEIVED DOSE PER YEAR [24]

*Source: ECRP (European Commission Radiation Protection) 112: Radiological protection principles concerning the natural radioactivity of building materials 1999 [25].

(ECRP) [25]. The activity index (*I*) should not exceed the values in table 2.

In this study, it was found that the value of $I \leq 1$, showing that this volcano ash could be used as bulk or superficial material without regulation and still in the acceptable limit [14]. In addition to these results, it would be interesting to determine the exposure of the γ radiation with radium equivalent activity which can be defined by equation 2 [14].

$$Ra_{eq} = A_{Ra} + 1.43\,A_{Th} + 0.077\,A_K \leq 370 \quad (2)$$

The obtained radium equivalent activity for volcano ash is 116 ± 1 Bq/kg, which is well lower the optimum limit of 370 Bq/kg.

Water Quality Analysis

The volcano ash is erupting until 2012, primarily from a 60 m (now up to 100 m) wide central crater. It is also being pumped into the Kali Porong River diversion channel as shown in figure 1, which then carries the mud to the ocean approximately 20 km to the east [6]. This effort is as an alternative to reduce the high quantity of volcano ash. However, the effect to the water in river and environment need to be analyzed. Table 4 shows the preliminary analytical results for water volcano ash compared with the standard of Environmental Quality (Sewage & Industrial Effluent) Regulations 1979 [22]. The results indicate that the quantity of mercury, tin, arsenic, chromium, manganese, nickel, and boron are within the allowed limitation standard. The contents of cadmium, lead and zinc are slightly higher than standard for both types A and B. Analysis also indicated that copper and iron are both present in levels that substantially exceed the limitation standard for type B. Of the major species present in the waters, cyanide and sulphide are both present in level that substantially exceed the standards for types A and B. High sulphide content is toxic to aquarium fish. As this water is brought to the surface for use, the unpleasant smelling hydrogen sulphide gas has released. Sulphide occurs naturally in crude petroleum, natural gas, volcanic gases, and hot springs. It can also result from bacterial breakdown of organic matter. However, Geoffrey et al. [6] stated that the high sulphide content does not appear to have any potential to generate acid drainage due to sulphide oxidation. From the results produced, further investigation is needed to understand the effects of the volcano ash on the aquatic ecosystems and physical sedimentation standpoint. Geoffrey et al. [6] also stated that this volcano ash has the potential to adversely affect the quality of surface or ground water sources for drinking water.

Microstructure Characterization of Volcano Ash

Volcano Ash Particle Size Analysis (PSA)

Particle size analysis was performed by dry method using Mastersizers 2000 Malvern Instruments Version 5.22 on a dried volcano ash. The mean particle size of the volcano ash is dominated by particles in the size of 121μm with specific surface area of 0.151 m^2/g. This size can be finer if the samples through the ball milled. Finer particle will give high surface area to react in geopolymerization process and affect the strength of geopolymer [26].

X-Ray Diffraction (XRD)

An x-ray diffraction (XRD) analysis of volcano ash is shown in figure 3. This volcano ash is almost dominated by quartz phase. This material exhibit a highest peak at 2 thetha where 2 thetha = 26.4° due to higher intensity of quartz (Q) revealed the silicon oxide. This statement also proved by the XRF results which shows that the higher contents of SiO_2 represent with high peak of intensity in XRD result. Volcano ash showed other five intense diffraction peaks at 2 thetha values of 21.0, 36.4, 39.5, 42.5 and 50.2°, which are associated with quartz (Q) [27]. The 2 thetha values of 19.7° and 24.0° showed unidentified element (X).

Scanning Electron Microscopic (SEM)

Microstructure of volcano ash was observed by SEM micrographs as shown in figure 4. Volcano ash has plate-like structure similar to kaolin. The particles observed as plate which combined layer by layer to form a structure. Furthermore, the structure of volcano ash shows more layer stick together to form the bigger structure due to the existence of water. The shape at high magnification of 5000x of volcano ash showed agglomerate of irregular shape like tissue texture that has been torn which depending on the crushed and blended process during sample preparation. The plate-like structure contributed to smaller surface area for geopolymerization process compared to fly ash which has sphere microstructure [28].

Fourier Transform Infrared Spectroscopy (FTIR)

Figure 5 shows the FTIR adsorption bands of the volcano ash. There was no specific reference for FTIR vibrational bands for volcano ash, so the table reference from study carried out by Lee and Deventer [29] were used which summarized for fly ash and kaolin. Volcano ash showed characteristic peaks at 2364 cm^{-1} - 3621 cm^{-1}, corresponding to the OH- stretching vibration. H-O-H stretching at 1634 cm^{-1} assigned to the weakly-bound water molecules, which are adsorbed on the surface or trapped

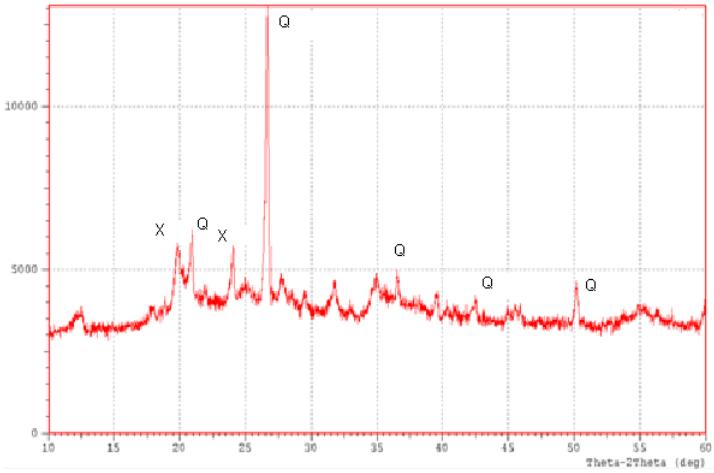

Fig. 3. X-ray diffractogram of volcano ash

Parameter	Units	Results		
		Volcano ash	Standard	
			A	B
Mercury as Hg	mg/l	0.0005	0.005	0.05
Tin as Sn	mg/l	<0.0004	0.2	1
Cadmium as Cd	mg/l	0.051	0.01	0.02
Arsenic as As	mg/l	<0.002	0.05	0.1
Lead as Pb	mg/l	0.769	0.1	0.5
Chromium as T.Cr	mg/l	0.071	0.2	1
Copper as Cu	mg/l	0.904	0.2	1
Manganese as Mn	mg/l	<0.03	0.2	1
Nickle as Ni	mg/l	ND (<0.01)	0.2	1
Zinc as Zn	mg/l	1.287	1	1
Iron as Fe	mg/l	2.504	1	5
Boron as B	mg/l	ND (<0.02)	1	4
Cyanide as CN^-	mg/l	16	0.05	0.1
Sulphide as $S2^-$	mg/l	80.60	0.5	0.5

Notes: "<" = Less than; "ND" = Not Detected;; "A" = Discharge upstream of water supply sources. "B" = Discharge downstream of water supply sources. Source: Environmental Quality (Sewage & Industrial Effluent) Regulations 1979 [22]

Table 4
PRELIMINARY ANALYTICAL RESULTS FOR WATER FROM VOLCANO ASH [22]

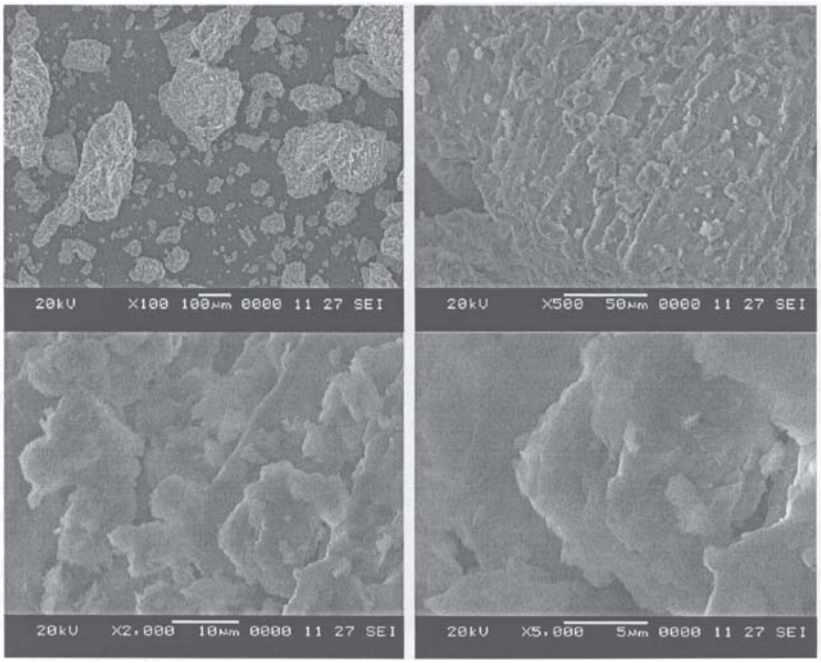

Fig. 4. SEM micrograph of volcano ash with various magnifications

in the large cavities between the agglomerated volcano ash. Band at 986 cm^{-1} was assigned to Si-O bonds in the SiO_4 molecules. Band at 910 cm^{-1} was attributed to Al-OH vibrations. The bands at 796 cm^{-1} and 777 cm^{-1} were Si-O-Si symmetric stretching. According to Fernandez-Jimenez and Palomo [30] the absorption peak at 781 cm^{-1} was an indication of the presence of quartz in kaolin. That means, the absorption peak at 796 cm^{-1} and 777 cm^{-1} was highly suggested as the presence of quartz in volcano ash as observed in XRD analysis. The bands at 749 cm^{-1} and 690 cm^{-1} were assigned to Al-O-Si kaolinite bending. These Si-O-Si and Si-O-Al vibrations may affect the strength of the structure. Table 5 summarized the FTIR absorption peaks for volcano ash.

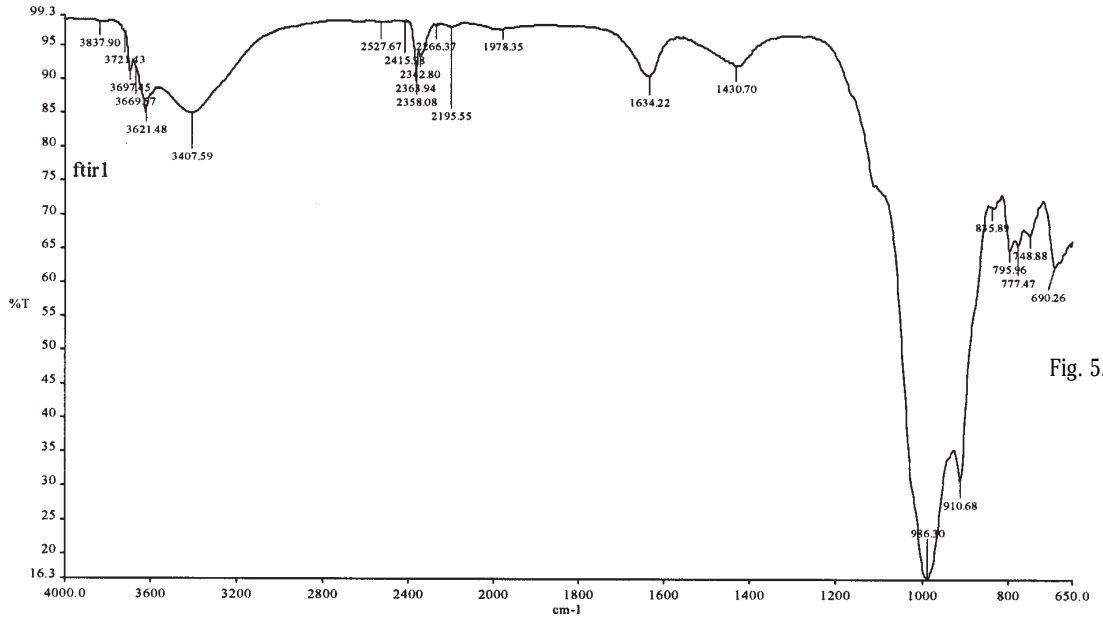

Fig. 5. FTIR spectra of volcano ash.

Bonds	Wave number (cm^{-1})
OH- stretching vibration	2364-3621
H-O-H stretching	1634
Si-O bonds	986
Al-OH vibrations	910
Si-O-Si symmetric stretching	796, 777
Al-O-Si kaolinite bending	690, 749

Table 5
MAIN FTIR ABSORPTION PEAKS OF VOLCANO ASH

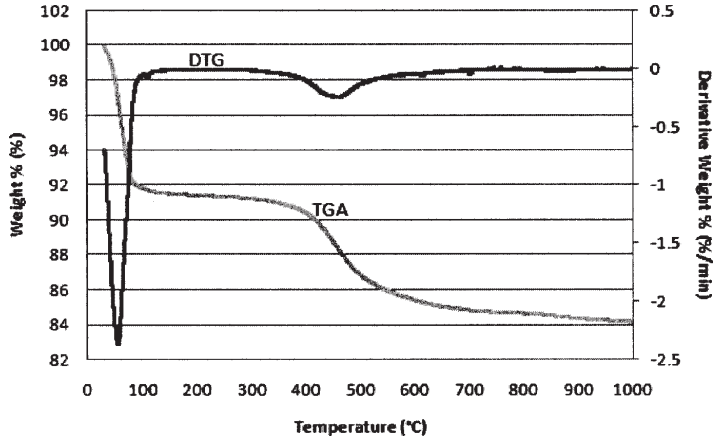

Fig. 6. TGA and DTG of volcano ash.

Thermogravimetric Analysis (TGA)/Differential Thermogravimetric Analysis (DTG)

Thermogravimetric analysis (TGA) was conducted to measure the weight loss while the specimens were gradually exposed to elevated temperature from 50°C to 1000°C at 5°C/min. Powdered sample were prepared to be used during testing. Figure 5 shows the differential thermogravimetric (DTG) and TGA curves for the volcano ash. The volcano ash experienced mass reduction with increasing temperature. Rapid rate of weight loss due to loss of water (volcano ash dominated by clay minerals), at 30-90°C and 430-500°C temperature ranges as recorded in the TGA curve. The average total percentage of weight remaining after being heated to 1000°C was 84.15%. The rate of weight loss stabilized between 510 and 1000°C. The decrease in weight peaked at 55°C as indicated by DTG curve.

Potential of Volcano Ash as Geopolymer Artificial Aggregate

Davidovits [31] has proposed the applications of geopolymeric materials based on silica-to-alumina atomic ratio. The Si/Al ratio for volcano ash found by XRF (table 1) was 2.74 (between 2-3) showing this material can be used as low CO_2 cements, concretes, radioactive and toxic waste encapsulation, heat resistance composites, foundry equipment and fibre glass composites.

Further study may explore the potential of volcano ash as building materials focusing on development of geopolymer artificial aggregate as an effort to explore the potential benefits of the unstoppable flow of volcano ash. On the other hand, the natural aggregate resource now is depleting day by day [32], however, the demand for aggregates in the market is large and increasing continuously. So, the alternative of producing a new geopolymer artificial aggregate with volcano ash with further research will utilize the natural resource [33] eventually producing a new geopolymer artificial aggregate with better properties.

Conclusions

As a conclusion from radioactive concentration of ^{226}Ra, ^{232}Th, ^{40}K, and ^{238}U were within the acceptable limits and much lower than study carried out by other researcher. The activity index, *I* value was less than 1, showing that

this volcano ash could be used as bulk or superficial material which were within the acceptable limit. The result also shows that the radium equivalent activity for volcano ash is 116 ± 1 Bq/kg, which is well lower the optimum limit of 370 Bq/kg. Water that is derived from the volcano ash may have adversely affected the aquatic ecosystem and human health which is not safe to be used as effluent. Further works need to be done on the toxicity test of this water from volcano ash.

The mean particle size of the volcano ash was 121 μm. The Si/Al of volcano ash is 2.74 and this material almost dominated by quartz phase. Microstructure analysis showed that volcano ash had a plate-like structure similar to kaolin. The FTIR adsorption band showed the OH-, H-O-H, Si-O, Al-OH, and Si-O-Si and Si-O-Al vibrations appeared in this volcano ash. The volcano ash experienced mass reduction with increasing temperature. The average total percentage of weight loss after being heated to 1000 °C was 15.85%. From the results obtained, it shows that volcano ash has a potential to be used as geopolymer raw material with acceptable limit for radiation. In the near future, this volcano ash can be used as raw material for geopolymer artificial aggregates instead of using natural resources materials with better properties.

References

1.MAZZINI, A., ETIOPE, G., SVENSEN, H., Earth and Planetary Science Letters, **317**, 2012, p. 305.

2.DAVIES, R., SWARBRICK, R., EVANS, R.J., HUUSE, M., GSA Today, **17**, 2, 2007. p. 4.

3.MAZZINI, A, SVENSEN, H, AKHMANOV, G.G., ALOISI, G., PLANKE S, Earth and Planetary Science Letters, **261**, 2007, p.375.

4.DAVIES, R.J., BRUMM, M., MANGA, M., RUBIANDINI, R., SWARBRICK, R., TINGAY, M., Earth and Planetary Science Letters, **272**, 2008, p. 627.

5.CRYANOSKI, D., Nature, **45**, 2007, p. 470.

6.PLUMLEE, G.S., HANDOKO, T.J.C., WIBOWO, T., ROSENBAUER, R.J., JOHNSON, C.A.,. BREIT, G.N., Preliminary Analytical Results for a Mud Sample Collected from the LUSI Mud Volcano, Sidoarjo, East Java, Indonesia. U.S. Geological Survey, Reston, Virginia, 2008, p. 1-26.

7.DAVIES, R., MANGA, M., TINGAY, M., SWARBRICK, R., Engineering Geology, **121**, 2011, p. 97.

8.NURUDDIN, M.F., BAYUAJI, R., MASILAMANI, M.B., BIYANTO, T.R., Civil Engineering Dimension, **12**, 1, 2010, p. 18.

9.DAVIDOVITS, J., Properties of Geopolymer Cements, Proceedings First International Conference on Alkaline Cements and Concretes, Kiev, Ukraine, Geopolymer Institute, Saint-Quentin, France, 1994. p. 131 - 149.

10.DAVIDOVITS, J., High-alkali cements for 21st century concretes. In Concrete Technology, Past, Present and Future, (Editor Metha, P.K.), American Concrete Institute, Farmington Hills, 1994.

11.DAVIDOVITS, J., DAVIDOVICS, M., Geopolymer: Ultra-High Temperature Tooling Material For the Manufacture of Advanced Composites, SAMPE Symposium and Exhibition, **36**, 1991, p. 1939.

12.DAVIDOVITS J., Journal of Thermal Analysis and Calorimetry, 35, 2, 1989, p. 429.

13.LLOYD, N.A., RANGAN, B.V., Geopolymer concrete with fly ash, Second International Conference on Sustainable Construction Materials and Technologies, Ancona, Italy, 2010.

14.RYUTA, H., KIYOSHI, S., Environment Asia, **2**, 2009, p. 45.

15.BOCIORT D., GHERASIMESCU, C., BERARIU, R., BUTNARU, R.,BRANZILA, M., SANDU, I., Rev Chim (Bucharest), **63**, no. 11, 2012, p. 1152

16.BOCIORT D., GHERASIMESCU, C., BERARIU, R., BUTNARU, R.,BRANZILA, M., SANDU, I., Rev Chim (Bucharest), **63**, no. 12, 2012, p. 1243

17.CALU, N., RANG, A., CECAL, A., BERDAN, I., SANDU, I., Rev. Chim.(Bucharest), **32**, no. 9, 1981, p. 896.

18.BEJINARIU, C., SANDU, A.V., BACIU, C., SANDU, I., TOMA, S.-L., SANDU, I.G., Rev. Chim. (Bucharest), **61**, no. 10, 2010, p. 961.

19.COPCIA, V., HRISTODOR, C., LUCHIAN, C., BILBA, N., SANDU, I., Rev. Chim. (Bucharest), **61**, no. 12, 2010, p. 1192.

20.I.K. KOUAME, B. DIDI, K. KOFFI, I. SAVANE, I. SANDU, Internationl Journal of Conservation Science, **1**, 3, 2010, p. 156.

21.J.M. SIELIECHI, G. J. KAYEM, I. SANDU, Internationl Journal of Conservation Science, 1, 3, 2010, p. 175.

22.*** Environmental Quality (Sewage & Industrial Effluent) Regulations 1979, Regulations 8(1), 8(2), 8(3), Parameter Limits of Effluents of Standards A and B, 1979

23.*** United Nations Disaster Assessment and Coordination (UNDAC), Environmental Assessment Hot Mud Flow East Java, Indonesia. Switzerland: UNEP/OCHA Environmental Unit, 2006.

24.SOMLAI, J., JOBBAGY, V., KOVACS, J., TARJAN, S., KOVACS, T., Journal of Hazardous Materials, **150**, 2008, p. 541.

25.*** European Commission Radiation Protection (ECRP), Radiological protection principles concerning the natural radioactivity of building materials, 1999. p. 1-16.

26.HARDJITO, D., WIBOWO, A.G.M., CHRISTIANTO, D., Materials, **5**, 9, 2012, p. 1654.

27.DUCHESNE, J., DUONG, L., BOSTROM, T., FROST, R., Waste and Biomass Valorization, **1,** 2010, p. 367.

28.MUSTAFA AL BAKRI, A.M., KAMARUDIN, H., BINHUSSAIN, M., NIZAR, I.K., RAFIZA, A.R., ZARINA, Y., Reviews on Advanced Materials Science, **30**, 2012, p. 90.

29.LEE, W.K.W., DEVENTER, J.S.J., Colloids and Surfaces A - Physicochemical and Engineering Aspects, **211**(2-3), 2002, p. 115.

30.FERNANDEZ-JIMENEZ, A, PALOMO, A., Micropor Mesopor Mater, **86**, 2005, p. 207.

31.DAVIDOVITS, J., Chemistry of Geopolymeric Systems, Geopolymer '99 International Conference, (Editors: Davidovits J., Davidovits R., James C.), 1999. p. 9-40.

32.DAVIDOVITS, J., World Resource Review, **6**, 1994, p. 263.

33.RAFIZA, A.R., MUSTAFA AL BAKRI, A.M., KAMARUDIN, H., KHAIRUL NIZAR, I., HARDJITO, D., ZARINA, Y., Advanced Materials Research, **626**, 2013,p. 892.

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